Fundamental Properties of Crystal-Melt Interfaces – Interface Moblity

Personnel: Ralph E. Napolitano (PI)

Abstract:

The intrinsic mobility of a solid-liquid interface is investigated by examining the interface response to uniaxial oscillation of the thermal field. Using transparent alloys of succinonitrile-acetone, the oscillation of the interface is observed directly. The frequency dependence of the amplitude and phase angle are related to the interfacial mobility. Preliminary experimental results are compared with a numerical model. Further development will lead to mobility measurements for various crystallographic orientations, enabling the assessment of kinetic anisotropy.

Background:

A general description of alloy solidification dynamics requires an understanding of interface response to the instantaneous local environment. Physical processes such as the diffusional redistribution of chemical species, the generation of interfacial curvature, and the attachment of atoms to the growing solid all require a driving free energy, which can be expressed as an interfacial undercooling,

$$\Delta T = \Delta T_D + \Delta T_R + \Delta T_K \tag{1}$$

where the subscripts indicate contributions from solute, curvature, and interface kinetics, respectively. Here, we are interested in the third term, which is typically very small for metallic systems, and is often neglected. Recent experimental and theoretical results, however, suggest that the interfacial mobility, particularly the anisotropy in the mobility, may play a critical role in the dynamics of growth and morphological selection.

Defining the interface mobility, μ, as a *linear compliance*, the kinetic contribution is given by

$$\Delta T_K = \frac{V}{\mu}. \tag{2}$$

For a pure metal, $\Delta T_D=0$, and the kinetic contribution is the only velocity-dependent term. In this case, the mobility is simply given by the linear dependence of the interface temperature on velocity. For alloys, the diffusional redistribution of solute is also dependent on the interface velocity, and experimental determination of mobility requires the separation of these effects. For this reason, mobility is best measured using a simple morphology, such as a plane front, where the diffusive effects are more easily accounted for. An additional complication is that metallic solid-liquid interfaces typically have very high mobility, and the corresponding kinetic undercoolings are very low. Accordingly, producing a measurable ΔT_K requires driving the interface at a relatively high velocity. This poses a two-fold problem for mobility measurement in alloys. Planar interfaces can only be maintained at very low velocities, and solutal transients are typically very long. In this work, an oscillating interface approach is used to measure the interfacial mobility. This method permits driving the interface at a high velocity without causing the destabilization of the plane front. The frequency response of such an oscillating system can be analyzed to determine the intrinsic mobility of the interface.

Theory, Modeling, and Experiment

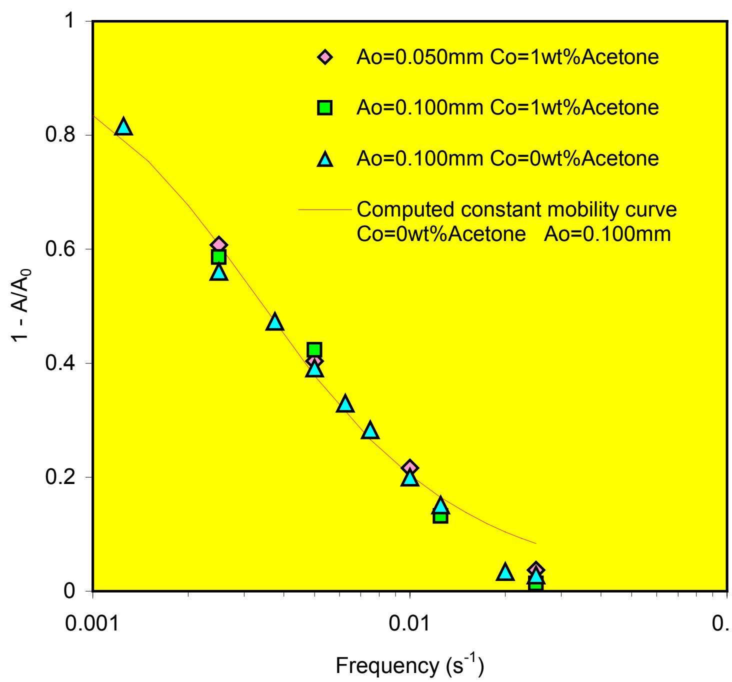
Both the independent motion of the oscillating thermal field and the dependent motion of the solid-liquid interface are defined as general 1-D Fourier modes out of phase by an angle, ϕ. The velocity of the thermal field, v_0 and the interface velocity, v^* are given by

$$v_0 = A_0 \omega \cos(\omega t) \qquad \text{and} \qquad v^* = A\omega \cos(\omega t - \phi)\,. \tag{3}$$

Noting that the interface undercooling is the product of the thermal gradient and the displacement within the gradient frame, we define a dimensionless mobility, $\mu' = \mu G/\omega$, and apply Eq.(2),

$$\mu' = \alpha \left[\frac{\cos(\omega t - \phi)}{\sin(\omega t) - \alpha \sin(\omega t - \phi)} \right] \tag{4}$$

where α is the normalized amplitude of interface oscillation, given by A/A_0.



The free boundary response to the oscillation of the thermal field is modeled numerically to reveal the expected self-similar response when motion is controlled only by interface kinetics. Employing an alternating constant velocity (i.e. saw-toothe wave), the frequency response of the normalized steady-state interface amplitude and can be predicted. Furthermore, in the fully kinetic limit, this response is independent of the driving amplitude and quite sensitive to the interface mobility, providing a means for experimental determination. Some preliminary experimental measurements are plotted in the figure to the right, showing reasonable agreement with a constant-μ numerical solution. No value of μ is reported here because the analysis of thermal relaxation is currently not yet complete.

Significance:

The mobility is a fundamental property of crystal-melt interfaces and therefore important in all solidification processes. The influence of interface kinetics on morphological selection is particularly important and is a primary area of investigation in the solidification community.

Future Work:

The analysis of the oscillating interface will be developed fully from both the theoretical and experimental perspectives. Specifically, the effects of solutal and thermal diffusion and crystal orientation will be described quantitatively.

Interactions:

This work is part of the collaborative efforts of the Computational Materials Science Network, sponsored by the U.S. Department of Energy.